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(54) Title: IMPROVED PCT FORMULATIONS CONTAINING HALOGENATED IMIDES, PHENOXY COMPOUND(S) AND REINFORCING FIBERS

(57) Abstract: This invention relates to a polymer composition comprised of: (A) a polyester comprising: (1) terephthalic acid in the amount of 85 to 100 mole % based on the mole percentages of the dicarboxylic acid component equaling a total of 100 mole %; (2) a glycol component comprising from about 60 to 100 mole % 1,4-cyclohexanedimethanol; (B) one or more halogenated imides; (C) one or more phenoxy compounds; and (D) reinforcing fiber.

IMPROVED PCT FORMULATIONS CONTAINING HALOGENATED IMIDES, PHENOXY COMPOUND(S) AND REINFORCING FIBERS

CROSS REFERENCE TO RELATED APPLICATIONS

This application claims priority upon provisional application Serial No. 60/155,744 filed September 23, 1999, and the 60/155,744 application is herein incorporated by this reference in its entirety.

FIELD OF THE INVENTION

This invention relates to an improvement in oven aging stability of a fiber reinforced, flame retardant (FR) poly(cyclohexylenedimethylene terephthalate) (PCT) formulation.

BACKGROUND OF THE INVENTION

Certain electronics must be soldered at high temperatures and require high heat deflection temperature (HDT) materials. Such materials typically have high melting points and thus must be melt processed at a relatively high temperatures. PCT is a high-melting polyester with a melting point of about 290°C and, consequently, is very challenging to formulate while retaining good molecular weight. In addition to good melt stability, good dimensional stability characteristics are important. Oven aging stability and flammability characteristics are also important.

United States Patent 4,837,254 discloses the use of PCT, a halogenated flame retardant, a mixture of elemental antimony metal and Sb_2O3, and Phenoxy. The invention is based on the synergistic effects of antimony metal and phenoxy, the combination of which is necessary to give good HDT. This patent mentions that phenoxy with other FR synergists as defined herein gives inadequate HDT in the cited system. High levels of antimony are shown.

United States Patent 5,428,086 discloses examples comprising PCT, poly(dibromophenylene oxide) and phenoxy resin. Neither HDT nor oven aging character is recognized. Phenoxy is considered inferior to epoxy.

United States Patent 3,962,174 discloses use of phenoxy in PBT or poly(propylene terephthalate).

United States Patent 4,008,199 discloses use of phenoxy, polyamide and phosphonate in PBT.

United States Patent 5,371,123 by Gallucci et al discloses use of glass fibers with specific coatings (olefin or epoxy) to improve the color of a polyester with a halogenated bis-imide flame retardant. This choice of glass fiber coating is not relevant to the current invention. No specific mention of stability or oven aging is given. Also, polyurethane coated glass is disclosed as being inferior.

BRIEF SUMMARY OF THE INVENTION

This invention relates to a polymer composition comprised of:

(A) a polyester comprising:

(1) terephthalic acid in the amount of 85 to 100 mole % based on the mole percentages of the dicarboxylic acid component equaling a total of 100 mole %.

(2) a glycol component comprising from about 60 to 100 mole % 1,4-cyclohexanedimethanol;

(B) one or more halogenated imides;

(C) one or more phenoxy compounds; and

(D) reinforcing fiber.

A preferred embodiment of this invention comprises one or more phosphorous based compounds and/or sodium antimonate.

The polymer composition of this invention has improved dimensional stability, improved oven aging stability, and improved flammability

characteristics that are particularly useful in high temperature electronics. Preferred embodiments also have improved melt stability.

DETAILED DESCRIPTION

This invention relates to an improvement in oven aging stability as well as melt stability of a fiber reinforced, flame retardant (FR) poly(cyclohexylenedimethylene terephthalate) (PCT) formulation.

Specifically, this formulation simultaneously achieves good oven aging stability (as demonstrated by property retention after oven aging at 190°C for 500 hours) and acceptable flammability characteristics. This is typified by UL-94 flammability testing. Also, the relative hazard or environmental load of the metallic synergist can be considered in evaluating the usefulness of the invention.

It is preferred in this invention that the polyester comprise 90 mole % or more of terephthalic acid based on the mole percentages of the dicarboxylic acid component of the polyester equaling a total of 100 mole %. By terephthalic acid, suitable synthetic equivalents, such as dimethyl terephthalate, are included.

The polyester useful in this invention comprises 0 to 15 mole %, preferably 0-10 mole %, of dicarboxylic acids other than terephthalic acid, based on the mole percentages of the dicarboxylic acid component of the polyester equaling a total of 100 mole %. The other dicarboxylic acids include, but are not limited to aromatic dicarboxylic acids preferably having 4 to 40 carbon atoms, more preferably, 8 to 14 carbon atoms; aliphatic dicarboxylic acids having, preferably 4 to 40 carbon atoms, more preferably, 4 to 12 carbon atoms; or cycloaliphatic dicarboxylic acids having 4 to 40 carbon atoms, more preferably, 8 to 12 carbon atoms..

Particularly preferred examples of other dicarboxylic acids useful in forming the copolyester of the invention include, but are not limited to, isophthalic acid, naphthalenedicarboxylic acid, cyclohexanedicarboxylic

acid, 1,4-cyclohexanediacetic acid, diphenyl-4,4'-dicarboxylic acid, naphthalenedicarboxylate, succinic acid, glutaric acid, adipic acid, azelaic acid, sebacic acid, and the like.

Of these, isophthalic acid, naphthalenedicarboxylic acid, cyclohexanedicarboxylic acid and naphthalenedicarboxyate are preferred, either singly or in combination.

When cyclohexanedicarboxylic acid is used as a comonomer in the context of the invention, trans-, cis-, or cis/trans mixtures may be used.

Any of the naphthalenedicarboxylic acid isomers or mixtures of isomers may be used. Some preferred naphthalenedicarboxylic acid isomers include 2,6-, 2,7- 1,4- and 1,5- isomers.

It should be understood that "dicarboxylic acids", includes the corresponding acid anhydrides, esters, and acid chlorides of these acids. In the acid component of this invention, the mole percentages of the acids of the polyester referred to herein equal a total of 100 mole %.

In the glycol component of this invention, the mole percentages of the glycols referred to herein equal a total of 100 mole %.

In the invention, it is preferred that the glycol component of the copolyester of the invention contain from about 80 to 100 mole %, preferably 90 to 100 mole %, of one of the isomers of 1,4-cyclohexanedimethanol.

Preferably, the copolyesters of this invention may be based on trans-, or cis/trans mixtures of 1,4-cyclohexanedimethanol. For example, a 30/70 cis/trans mixture of the isomers may be readily used.

The glycol component may comprise up to 20 mole %, and more preferably, up to 10 mole %, of one or more other aliphatic or alicyclic glycols.

Such additional diols include cycloaliphatic diols preferably having 6 to 20 carbon atoms or aliphatic diols preferably having 2 to 20 carbon atoms. Examples of such diols are: ethylene glycol, diethylene glycol,

triethylene glycol, propane-1,3-diol, butane-1,4-diol, pentane-1,5-diol, hexane-1,6-diol, 3-methylpentanediol-(2,4), 2-methylpentanediol-(1,4), 2,2,4-trimethylpentane-diol-(1,3), 2-ethylhexanediol-(1,3), 2,2-diethylpropane-diol-(1,3), hexanediol-(1,3), 1,4-di-(hydroxyethoxy)-benzene, 2,2-bis-(4-hydroxycyclohexyl)-propane, 2,4-dihydroxy-1,1,3,3-tetramethyl-cyclobutane, 2,2-bis-(3-hydroxyethoxyphenyl)-propane, decalin diol and 2,2-bis-(4-hydroxypropoxyphenyl)-propane. Copolyesters may be prepared from the above diols in addition to the 1,4-cyclohexanedimethanol.

It is more preferred that the one or more glycols are selected from ethylene glycol, diethylene glycol, triethylene glycol, propanediol, butanediol, pentanediol, hexanediol, and tetramethylcyclobutanediol.

When the copolyester contains ethylene glycol, it is preferable that the ethylene glycol be present in an amount less than 20 mole %, more preferably, less than 10 mole %.

Copolyesters containing substantially only 1,4-cyclohexanedimethanol and terephthalic acid or substantially only 1,4-cyclohexanedimethanol, isophthalic and terephthalic acid are preferred.

The polyester resins useful in the blend of this invention are well known and are commercially available. By the term "polyester", copolyesters are also intended. Methods for their preparation are described, for example, in United States Patents 2,465,319 and 3,047,539. For example, the polyesters can be prepared by direct condensation of terephthalic acid or ester interchange using dimethyl terephthalate with the selected glycol.

Typical catalysts which may be used to make these copolyesters include titanium alkoxides, dibutyl tin dilaurate, combinations of zinc, manganese, or magnesium acetates or benzoates with antimony oxide or antimony triacetate.

The polyesters of the invention preferably have an inherent viscosity of 0.1 to 2.0 dL/g, more preferably 0.3 to 1.5 dL/g, and even more

preferably, 0.4 to 1.2 dL/g as measured at a temperature of 25°C for a 0.5 gram sample in 100 ml of a solvent consisting of 60% by weight phenol and 40% by weight tetrachloroethane.

It is preferred that the copolyester useful herein have a melting point of more than 260°C, more preferably, greater than 270°C. Preferably, the copolyester has a melting point of 260°C to 310°C.

Melting points for the polyester resins and halogenated imides of the invention are measured by DSC (differential scanning calorimetry) analysis.

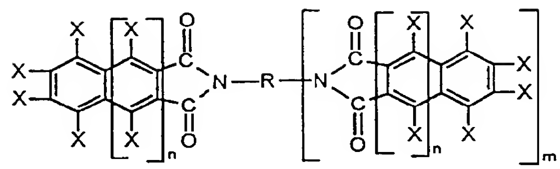
The invention is meant to incorporate all melt processing methods known in the art.

As used herein, the term "melt processing" refers to any processing step commonly used in the art for polyesters or copolyesters which occurs after the polyesters or copolyesters are heated to their melting temperature or melting point. This includes, but is not limited to, injection molding, calendering, extrusion and rotational molding.

It is also preferred within the context of this invention that the polymer of the invention undergoes less than a 50%, preferably 25%, and more preferably 15% loss in number average molecular weight as determined by gel permeation chromatography when the polymer composition is melt processed at 25°C above the crystalline melting temperature of the polymer composition (Tm) for 10 minutes.

The second component of the composition is an organic compound containing at least one imide group and having a melting point greater than 240°C. Useful families of imide-group containing compounds include N,N'-arylenediphthalimides wherein the arylene group includes phenylene, diphenylene, naphthylene and sulfone bridged bisphenyls, tetrabrominated phthalimides, N,N'bis(dibromocyclohexane dicarboxyimides) with various bridging groups, and N,N'-alkylenebis(tetrahalophthalimides).

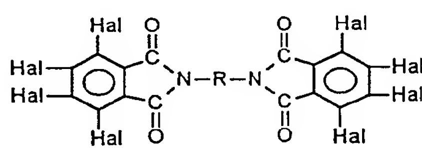
Preferred imide group containing compounds are those corresponding to the following formula:



wherein both n and m may be 1 or 0, X may be halogen, particularly chlorine or bromine, or hydrogen, and

R is a C_1 to C_6 alkyl group, a single bond, a phenylene group, a toluene group, a cyclohexylene group, a bis phenyl methane group, a bis cyclohexylmethane group, or a naphthylene group.

The N,N'-alkylenebis(tetrahalophthalimides) suitable in the present invention and a process for their production are described in U.S. Pat. No. 4,087,441, incorporated herein by reference. The preferred N,N'-alkylenebis(tetrahalophthalimides) are represented by the formula:



wherein R represents a C_1-C_6 alkyl group, preferably a C_2-C_6 alkyl group, and most preferably an ethyl group, and

Hal which may be the same or different, represents a halogen atom, preferably Br or Cl, and most preferably Br.

The most particularly preferred N,N'-alkylenebis(tetrahalophthalimide) is N,N'-ethylenebis(tetrabromophthalimide) (R is an ethyl group and Hal is a Br atom). These types of imide group containing components are described in United States Patent Nos. 3,624,024 and 3,873,567 and British Pat. No. 1,287,934.

Other suitable imide group containing compounds include 1,4,5,6-tetrabromo-2,3-phthaloimide; N methylol-tetrabromophthalimide; N,N-bis-(1,4,5,6-tetrabromo-2,3-phthaloimide); N,N'-p-phenylene-diphthalimide; N,N'-di-phthalimidodiphenyl; bis-(N phenyl-phthalimido)sulphone; N,N'-p-phenylene-di-tetrachlorophthalimide; 4,4'-di-tetrachlorophthalimidodiphenyl; N-(tetrachlorophthalimido)-tetrachlorophthalimide, N,N'-p-phenylene-di-tetrabromophthalimide; N,N'-di-tetrabromophthalimidodiphenyl; N-(tetrabromophthalimido)-tetrabromophthalimide; N,N'-bis-(5,6-dibromocyclohexane-2,3-dicarboximide); and N,N'-(1,2ethane)-bis-(5,6 dibromocyclohexane-2,3-dicarboximide).

Further suitable imide containing compounds are disclosed in U.S. Pat. Nos 3,868,388; 3,873,567; 3,915,930; 3,923,734; 4,001,179 and 4,003,862. Suitable imides are also disclosed in British Pat. No. 1,287,934 and are incorporated herein by reference.

Preferred imides have a melting point above 240°C, preferably above 300°C, which including bis-imides made from aromatic or aliphatic diamines, including ethylene diamine, or hydrazine, and tetrabromophthalic anhydride or acid are preferred. Again, the most preferred flame retardant is the imide from reacting tetrabromo phthalic acid (anhydride) with ethylene diamine. This is sold commercially as Saytex BT-93 and BT-93W. This flame retardant has a high bromine content. It is thermally stable to processing temperatures characteristic of PCT, and does not soften below the PCT melting point. This class of phthalimides has an advantage over other high temperature bromine sources like decabromodiphenyl in that they are not singled out as having the same environmental concerns (dioxins/furans).

It is preferred that the sum of all flame retardants used in this invention is 5-30%, preferably 10-20%, by weight of the total composition. One or more flame retardants may be used within the context of this invention.

Brominated phthalimides are high melting materials, in contrast to other brominated flame retardants, and provide superior HDT. However, the combination of PCT and BPI is especially challenging from a melt stability standpoint. The preferred FR synergists with PCT/BPI system are not the same as with lower melting point materials like PBT (Tm=220°C) or PET (Tm=250°C).

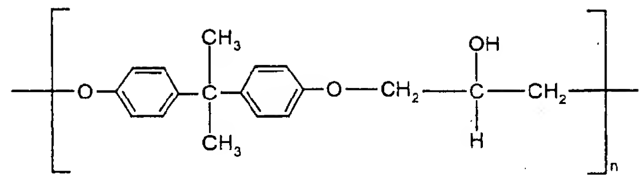
A number of patents describe processes for preparing brominated phthalimides, for example, United States Patents 4,997,953; 5,076,970; 5,290,945; 5,137,948; and 5,317,048, all of which are incorporated herein by reference.

Certain preferred polymer compositions of this invention achieve good dimensional stability and good melt stability due, at least in part, to use of the flame retardant synergist. Antimony compounds are preferred. Sodium antimonate is more preferred. Even more preferred is $NaSbO_3$ that is substantially free of Sb^{+3} or less than 1 mole % of Sb^{+3} based on the total mole percentages of antimony in the sodium antimonate.

Sodium antimonate is normally described in the art as being a sodium-neutralized version of Sb_2O_3 but the importance of Sb^{+3} level and the desire to reduce rather than increase the amount of synergist suggest this different mechanism, reducing catalytic activity of the Sb species.

It is even more preferred that the ratio of flame retardant (halogenated imides, preferably, brominated phthalimide(s)) to flame retardant synergist (sodium antimonate) is optimized at a much higher weight ratio (5:1 to 10:1, preferably 8:1) than is commonly practiced in the art (3:1 to 4:1).

The phenoxy compounds of the invention comprise a diepoxide monomer having the structural formula:

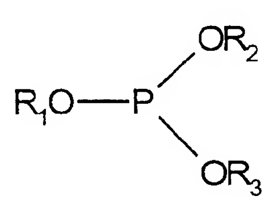


wherein n is from 10 to 100.

Examples of phenoxy compounds include the polyhydroxyether of bisphenol A which is produced from 2,2'-bis(4-hydroxy-phenyl) propane and epichlorohydrin. Preparation of such polymers is described in U.S. Patent No. 3,356,646.

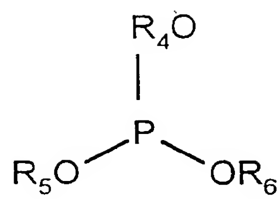
It is preferred that the phenoxy compounds of the invention comprise 1 to 10%, preferably 2 to 5% by weight of the total composition. It is also preferred that the phenoxy compound(s) of the invention have an Mn of 10,000 to 20,000.

It can also be shown that melt stability of the composition of the invention can be further improved by addition of a phosphorous-based compound. The phosphorous-based compounds include, but are not limited to, one or more phosphites or phosphonites wherein at least one of the P-O bonds is attached to an aryl radical. Such compounds may be represented by the formulas



Phosphonite

where at least one of R_1, R_2 and R_3 is an aryl radical of 6 to 30 carbon atoms and any other(s) of R_1, R_2 and R_3 are H or alkyl of 1 to 30 carbon atoms, or



Phosphonite

where at least one of R_4, R_5, and R_6 is an aryl radical of 6 to 30 carbon atoms.

Phosphites are preferred within the context of this invention.

Even more preferred are, for example, commonly available symmetrical triaryl esters of phosphorous acid which may be used are triphenyl phosphite; tris(nonylphenyl) phosphite; and tris(2,4-di-t-butylphenyl) phosphite (Irgafos 168). The most preferred symmetrical ester of phosphorous acid is bis(2,4-di-t-butylphenyl) pentaerythritol diphosphite (Ultranox® 626 - a trademark of General Electric Co.).

Examples of phosphorous-based compounds which may be used include, but are not limited to, Ultranox® 633 phosphite, (General Electric Chemicals), Irgafos®168 phosphite (Ciba-Geigy Corporation), Ethanox® 398 phosphonite (Ethyl Corporation) and Sandostab® P-EPQ phosphonite (Sandoz Chemicals).

The phosphorous compounds may be functioning by inhibiting the metal catalyst system present in the PCT. While much is known about use of phosphorous compounds in stabilizing polyester systems, it is not generally recognized as a way to stabilize flame retardants.

Without limiting the invention, it is suggested that all these changes may function by reducing the catalytic activity of the blend to a much lower level than is required for lower melting polyesters. This is in contrast to the usual concern of buffering the system from acidic HBr release which can directly attack the polyester.

It is understood that other additives such as other stabilizers, other flame retardants (FR), flame retardant synergists, tougheners, epoxy compounds, branching agents, mold release agents, nucleating agents, reinforcing agents such as glass fibers, carbon fibers, fillers, antioxidants, and colorants, such as carbon black, may also be desirable in such formulations. These may be added either during or after polymerization depending on the chemical structure of the addition.

Such additives are generally present at 0.1 to about 40 weight %, preferably 0.1 to about 20 weight %, based on the total weight of the copolyester composition.

Examples of reinforcing agents are glass fibers, carbon fibers, mica, clay, talc, wollastonite, and calcium carbonate. A particularly preferred reinforcing agent is glass fiber. It is preferable that the glass fibers be present in the polyester composition at from 0.1 to 45%, preferably 10 to 40%, by weight based on the total weight of said polyester composition.

Glass fibers suitable for use in the polyester compositions of the invention may be in the form of glass filaments, threads, fibers, or whiskers, etc., and may vary in length from about 1/8 inch to about 2 inches. Chopped glass strands having a length of about 1/8 inch to about 1/4 inch are preferred. Such glass fibers are well known in the art. Of course, the size of these glass fibers may be greatly diminished depending on the blending means employed, even to lengths of 300 to 700 microns or lower.

It is preferred that the glass fibers are coated with polyurethane.

The polyester compositions of the invention may be reinforced with a mixture of glass and other reinforcing agents as described above, such as mica or talc, and/or with other additives.

The components of the copolyester composition of the invention may be blended and/or mixed by any suitable technology known in the art. Compounding temperatures must be at least the melting point of the polyester. For example, the polyester can be mixed dry in any suitable

blender or tumbler with the other components and the mixture melt-extruded. The extrudate can be chopped. If desired the reinforcing material can be omitted initially and added after the first melt extrusion, and the resulting mixture can then be melt extruded.

The copolyester of this invention may be melt processed and extruded, injection molded or compression molded into a variety of shapes and forms including fibers, molded parts, bottles, pellets, containers, sheeting, film and the like. The product is especially suitable as an injection molding material for producing molded articles.

Unless otherwise specified, all parts, percentages, ratios, etc., are by weight. Weight of reinforcing glass fibers is based on total composition weight.

This invention can be further illustrated by the following examples of preferred embodiments thereof, although it will be understood that these examples are included merely for purposes of illustration and are not intended to limit the scope of the invention unless otherwise specifically indicated. The starting materials are commercially available unless otherwise indicated.

The following definitions of terms are applicable throughout this invention unless otherwise specified:

Abbreviations for the tables are as follows; other abbreviations and definitions may otherwise appear herein:

Flame Retardant 1 – (FR1) - N,N'-ethylenebis(tetrabromophthalimide) which is reaction product of ethylene diamine and tetrabromophthalic anhydride;

Flame Retardant 2 – (FR2) - brominated polystyrene;

Flame Retardant Synergist – (FRS1) - sodium antimonate with less than 1 mole % Sb^{+3};

Glass Fibers Type 1 – (GF1) - 11 microns, coupled with amino silane;

Glass Fibers Type 2 – (GF2) - 10 microns diameter, amino silane coupler, and thermoplastic polyurethane coating;

Glass Fibers Type 3 – (GF3) - (with 10 micron diameter, amino silane coupler, and thermoplastic polyurethane coating;

Glass Fibers Type 4 – (GF4) - 10 microns, amino silane, relatively free of coupler;

GPC - gel permeation chromatography;

PCT - poly(cyclohexylenedimethylene terephthalate);

Plasticizer 1 – (PL A) - polyethylene glycol dilaurate;

Stabilizer 1 – (ST1) - epoxy cresol novolac resin;

Stabilizer 2 – (ST2) - polymer of tris(4-glycidyloxyphenyl) methane;

Stabilizer 3 – (ST3) - tetrakis -[methylene -(3,5-di-tert-butyl-4-hydroxyhydro-cynnamate)];

Stabilizer 4 – (ST4) - [bis(2,4-di-t-butylphenyl)pentaerythritol diphosphite;

Stabilizer 5 – (ST5) - Phenoxy PKHH sold by Phenoxy Associates, Inc.

EXAMPLES

The fiber reinforced PCT blends of this work were prepared by extrusion compounding using a twin screw extruder at temperatures of 305°C. The resulting pellets were injection molded into tensile and flexural bars for use in mechanical and flammability property testing. Melt stability was determined on these blends by drying a small sample of the compounded pellets. Adequate drying can be obtained by drying in a vacuum oven overnight at 80°C, or by drying for 4 hours at 125°C in a hot air circulating oven. The dried pellets were then loaded into a Tinius Olsen melt indexer or capillary rheometer and held for 10 minutes at 305°C melt temperature, then analyzed by gel permeation chromatography. The melt

stability of these blends was shown by the retention of number average molecular weight (Mn) and weight average molecular weight (Mw) after 10 minutes at 305°C.

Oven Aging Stability - Oven aging stability was determined by comparing physical properties of molded test bars before and after aging in a circulating hot air oven at 190°C for 500 hours. Good aging stability is characterized by no worse than -25% change, preferably no change or even a positive value, indicating an increase in physical properties on aging.

The blends in the Examples below are described as follows. Percentages are by weight of the total composition.

EXAMPLE 1

Blends A-B in Table 1 are described as follows:

Blend A is described as follows:

30.0% GF1
14.0% FR2
3.5% FRS1
4.0% PL-A
0.5% ST2
0.5% polyethylene wax
0.25% ST3
0.25% ST4, and

PCT in an amount sufficient for the composition to total 100 weight%.

Blend B is described as follows:

Same as Blend A except that FR2 is replaced by 10.0% FR1

Table 1

	Blend A	Blend B
Flammability Rating	V0	V2
HDT @1.8 Mpa,°C	253	261
Tensile Strength, J/m	118	113
% Change on Aging	-28	-26

Table 1 shows that halogenated imides (for example, brominated phthalimides) improve heat deflection temperature with little impact on oven aging when phenoxy isn't used.

EXAMPLE 2

Blends C-D in Table 2 are described as follows:

Blend C is described as follows:

30.0% GF3
14.0% FR2
3.5% FRS1
4.0% PL-A
0.5% ST1
0.5% polyethylene wax
0.25% ST3
0.25% ST4, and

PCT in an amount sufficient for the composition to total 100 weight%.

Blend D is described as follows:

Same as Blend A except that ST1 is replaced by 3.0% ST5.

Table 2

	Blend C	Blend D
Flammability Rating @ 1/16"	V0	V0
HDT @1.8 Mpa,°C	259	258
Tensile Strength, J/m	113	114
% Change on Aging	-29	-18

Table 2 shows that phenoxy (ST5) aids oven aging properties only slightly when halogenated imides (for example, brominated phalimides) are not used and does not substantially harm heat deflection temperature.

EXAMPLE 3

Blends E-H in Table 3 are described as follows:

Blend E is described as follows:

30.0% GF2
16.0% FR1
2.0% FRS
4.0% PL-A
1.0% ST1
0.5% polyethylene wax
0.25% ST3
0.25% ST4, and

PCT in an amount sufficient for the composition to total 100 weight%.

Blend F is described as follows:
Same as Blend E except that epoxy is replaced by 3.0% ST5.

Blend G is described as follows:
Same as Blend E except that GF2 is changed to GF4.

Blend H is described as follows:
Same as Blend G except that ST1 is replaced by 3.0% ST5.

Table 3

	Blend E	Blend F	Blend G	Blend H
Flammability Rating @ 1/16"	V0	V0	V0	V0
HDT @1.8 Mpa,°C	263	261	262	260
Tensile Strength, J/m	116	94	107	96
% Change on Aging	-32	+4	-26	-4

Table 3 shows that phenoxy (ST5) dramatically improves oven aging properties when halogenated imides (example, brominated phthalimides) are used and that the effect is even stronger with polyurethane-coated glass fibers.

EXAMPLE 4

Blends I-M in Table 4 are described as follows:

Blend I is described as follows:

30.0% GF2
16.0% FR2
2.0% FRS
4.0% PL-A
1.5% ST1
0.5% polyethylene wax
0.25% ST3
0.25% ST4, and

PCT in an amount sufficient for the composition to total 100 weight%.

Blend J is described as follows:
Same as Blend I except that epoxy (ST1) is reduced to 1.0%.

Blend K is described as follows:
Same as Blend J except that 3.0% phenoxy (ST5) is added.

Blend L is described as follows:
Same as Blend K except that ST1 is completely eliminated.

Blend M is described as follows:
Same as Blend G except that Phenoxy (ST5) is increased to 4.5%

Table 4

Blends	Blend I	Blend J	Blend K	Blend L	Blend M
% Multifunctional Epoxy (ST1)	1.5	1	1	0	0
% Phenoxy (ST5)	0	0	3	3	4.5
Flammability Rating, 1/16" (as molded)	V0	V0	V0	V0	V0
HDT @1.8 Mpa,°C	257	259	255	259	256
Tensile Strength, J/m	117	118	116	101	103
% Change on Aging	-28	-29	-11	+2	+5

Table 4 shows that phenoxy (ST5) dramatically improves oven aging properties in the presence of halogenated imides (example, brominated phthalimides). Also, addition of an epoxy compound (ST1) interferes with the effect of the phenoxy compound (ST5) so that oven aging properties actually are worsened.

The invention has been described in detail with particular reference to preferred embodiments thereof, but it will be understood that variations and modifications can be effected within the spirit and scope of the invention.

We claim:

1. A polymer composition comprised of:
   (A) a polyester comprising:
      (1) terephthalic acid in the amount of 85 to 100 mole % based on the mole percentages of the dicarboxylic acid component equaling a total of 100 mole %.
      (2) a glycol component comprising from about 60 to 100 mole % 1,4-cyclohexanedimethanol;
   (B) one or more halogenated imides;
   (C) one or more phenoxy compounds; and
   (D) reinforcing fiber.

2. The composition of Claim 1 wherein terephthalic acid is present in the amount of 90 to 100 mole %.

3. The composition of Claim 1 wherein said dicarboxylic acid component comprises repeat units of from 0 to 15 mole % or less of one or more other dicarboxylic acids

4. The composition of Claim 3 wherein said one or more dicarboxylic acids is selected from cyclohexanedicarboxylic acid, isophthalic acid, 1,4-cyclohexanediacetic acid, diphenyl-4,4'-dicarboxylic acid, succinic acid, glutaric acid, adipic acid, azelaic acid, naphthalenedicarboxylic acid, and sebacic acid.

5. The composition of Claim 4 wherein said one or more dicarboxylic acids is selected from isophthalic acid, naphthalenedicarboxylic acid, cyclohexanedicarboxylic acid and naphthalene dicarboxylate.

6. The composition of Claim 5 wherein said one or more dicarboxylic acid is selected from isophthalic acid and naphthalenedicarboxylic acid.

7. The composition of Claim 6 wherein said dicarboxylic acid comprises isophthalic acid.

8. The composition of Claim 5 wherein said acid component comprises cyclohexanedicarboxylic acid.

9. The composition of Claim 5 wherein said acid component comprises naphthalenedicarboxylic acid.

10. The composition of Claim 1 wherein said glycol component comprises 80 to 100 mole % 1,4-cyclohexanedimethanol.

11. The composition of Claim 10 wherein said glycol component comprises 90 to 100 mole % 1,4-cyclohexanedimethanol.

12. The composition of Claim 10 wherein said glycol component comprises up to 20 mole % of one or more other aliphatic or alicyclic glycols.

13. The composition of Claim 12 wherein said glycol component comprises up to 10 mole % of one or more other aliphatic or alicyclic glycols.

14. The composition of Claim 13 wherein said one or more other glycols is selected from the group consisting of cycloaliphatic diols having 6 to 20 carbon atoms and aliphatic diols having 2 to 20 carbon atoms.

15. The composition of Claim 14 wherein said one or more other glycols is selected from the group consisting of ethylene glycol, diethylene glycol, triethylene glycol, propane-1,3-diol, butane-1,4-diol, pentane-1,5-diol, hexane-1,6-diol, 3-methylpentanediol-(2,4), 2-methylpentanediol-(1,4), 2,2,4-trimethylpentane-diol-(1,3), 2-ethylhexanediol-(1,3), 2,2-diethylpropane-diol-(1,3), hexanediol-(1,3), 1,4-di-(hydroxyethoxy)-benzene, 2,2-bis-(4-hydroxycyclohexyl)-propane, 2,4-dihydroxy-1,1,3,3-tetramethyl-cyclobutane, 2,2-bis-(3-hydroxyethoxyphenyl)-propane, decalin diol and 2,2-bis-(4-hydroxypropoxyphenyl)-propane.

16. The composition of Claim 15 wherein said one or more other glycols is selected from the group consisting of ethylene glycol, diethylene glycol, triethylene glycol, propanediol, butanediol, pentanediol, hexanediol, and tetramethylcyclobutanediol.

17. The composition of Claim 15 wherein said one or more other glycols comprises ethylene glycol in an amount less than 20 mole %.

18. The composition of Claim 15 wherein said one or more other glycols comprises ethylene glycol in an amount less than 10 mole %.

19. The composition of Claim 1 wherein said copolyester has a melting point of 260°C or more.

20. The composition of Claim 19 wherein said copolyester has a melting point of 270°C or more.

21. The composition of Claim 19 wherein said copolyester has a melting point of 260°C to 310°C.

22. The composition of Claim 1 wherein said polymer composition undergoes less than a 50% loss in number average molecular weight as determined by gel permeation chromatography when melt processed at about 25°C above the crystalline melting temperature for 10 minutes.

23. The composition of Claim 22 wherein said polymer composition undergoes less than a 25% loss in number average molecular weight as determined by gel permeation chromatography when melt processed at about 25°C above the crystalline melting temperature for 10 minutes.

24. The composition of Claim 1 comprising one or more additives selected from the group consisting of stabilizers, flame retardants, flame retardant synergists, tougheners, epoxy compounds, mold release agents, nucleating agents, reinforcing agents and branching agents.

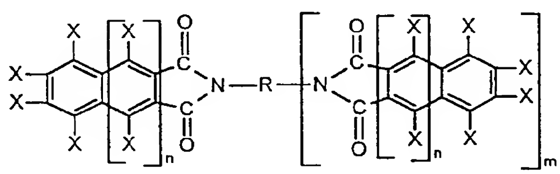
25. The composition of Claim 1 wherein said organic compounds containing at least one imide group have a melting point above 240°C.

26. The composition of Claim 1 wherein said organic compounds containing at least one imide group have a melting point above 300°C.

27. The composition of Claim 25 wherein said organic compounds containing at least one imide group comprise N,N'-arylenediphthalimides wherein the arylene group includes phenylene, diphenylene, naphthylene and sulfone bridged bisphenyls; tetrabrominated phthalimides; N,N'bis(dibromocyclohexanedicarboxyimides) with bridging groups; and N,N'-alkylenebis(tetrahalophthalimides).

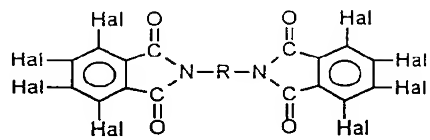
28. The composition of Claim 27 wherein said imide group containing compounds have the following formula:

wherein both n and m are either 1 or 0; X is selected from halogen, and hydrogen, and



R is selected from the group consisting of one or more of a C_1 to C_6 alkyl group, a single bond, a phenylene group, a toluene group, a cyclohexylene group, a bis phenyl methane group, a bis cyclohexylmethane group, and a naphthylene group.

29. The composition of Claim 27 wherein said imide group containing compounds are N,N'-alkylenebis(tetrahalophthalimides) having the formula:



wherein R is a C_1-C_6 alkyl group, and Hal represents a halogen atom.

30. The composition of Claims 28 or 29 wherein said halogen atom is selected from the group consisting of chlorine and bromine.

31. The composition of Claim 30 wherein said halogen atom is bromine.

32. The composition of Claim 29 wherein R is ethyl.

33. The composition of Claim 29 wherein said imide group containing compounds are brominated phthalimides made from aromatic or aliphatic diamines and from tetrabromophthalic anhydride or tetrabromophthalic acid.

34. The composition of Claim 33 wherein said diamines are selected from the group consisting of ethylene diamine and hydrazine.

35. The composition of Claim 33 wherein said imide group containing compound is N, N'-ethylenebis(tetrabromophthalimide).

36. The composition of Claim 24 wherein said flame retardant synergist is sodium antimonate.

37. The composition of Claim 1 wherein said sodium antimonate contains less than 1 mole percent of Sb+3 based on the total mole % of antimony present in the sodium antimonate.

38. The composition Claim 1 wherein the amount of brominated phthalimide(s) and sodium antimonate in the composition results in a weight ratio of 5:1 to 10:1.

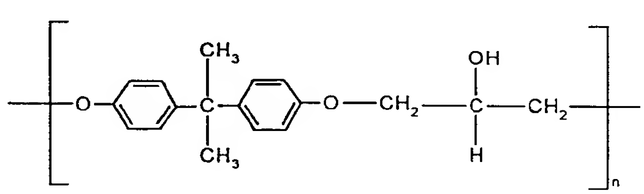
39. The composition of Claim 38 wherein the amount of brominated phthalimide(s) and sodium antimonate present in said composition results in a weight ratio of 8:1 of brominated phthalimide(s):sodium antimonate.

40. The composition of Claim 1 comprising 1 to 10% by weight of phenoxy compound(s) based on the weight of the total composition.

41. The composition of Claim 41 comprising 2 to 5% by weight of phenoxy compound(s) based on the weight of the total composition.

42. The composition of Claim 1 wherein said one or more phenoxy compounds have an Mn of 10,000 to 20,000.

43. The composition of Claim 1 wherein said phenoxy compound comprises the structural formula:



wherein n is 10 to 100.

44. The composition of Claim 43 wherein said phenoxy compound is the polyhydroxyether of bisphenol A which is produced from 2,2'-bis(4-hydroxy-phenyl) propane and epichlorohydrin.

45. The composition of Claim 24 additionally comprising one or more phosphorous based compounds.

46. The composition of Claim 45 wherein said phosphorous based compounds are selected from the group consisting of phosphites and phosphonites.

47. The composition of Claim 46 wherein said phosphorous based compounds are one or more phosphites.

48. The composition of Claim 47 wherein said one or more phosphites is bis(2,4-di-t-butylphenyl)pentaerythritol diphosphite.

49. The composition of Claim 24 wherein said reinforcing agents are selected from the group consisting of glass fibers, carbon fibers, mica, clay, talc, wollastonite, and calcium carbonate.

50. The composition of Claim 1 wherein said reinforcing fiber is selected from the group consisting of carbon fibers and glass fibers.

51. The composition of Claim 50 wherein said reinforcing agent is glass fiber.

52. The composition of Claim 51 wherein said glass fibers are present at from 0.1 to 45% by weight based on the total weight of the polymer composition.

53. The composition of Claim 51 wherein said glass fibers are chopped glass strands having a length of from about 1/8 inch to about 2 inches.

54. The composition of Claim 52 wherein said glass fibers are chopped glass strands having a length of from about 1/8 inch to about 1/4 inch.

55. The composition of Claim 51 wherein said glass fibers are coated with polyurethane

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 00/24378

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C08K13/04 C08L67/02 //(C08K13/04,5:3417,7:02)

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C08K C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, EPO-Internal, PAJ, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 604 078 A (GEN ELECTRIC) 29 June 1994 (1994-06-29)	1,10,24, 25, 27-35, 50,51
	page 2, line 50 -page 3, line 15; claims 1,7,8,1-3,14,24; examples; tables page 8, line 1-46	
A	PATENT ABSTRACTS OF JAPAN vol. 018, no. 313 (C-1212), 15 June 1994 (1994-06-15) & JP 06 065482 A (UNITIKA LTD), 8 March 1994 (1994-03-08) abstract	1,10,38
A	US 5 011 877 A (MORRIS JOHN C ET AL) 30 April 1991 (1991-04-30) column 3, line 3 - line 57	1,4-11, 24

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
11 January 2001	22/01/2001
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Authorized officer Engel, S

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Inten nal Application No
PCT/US 00/24378

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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A	PATENT ABSTRACTS OF JAPAN vol. 018, no. 686 (C-1292), 26 December 1994 (1994-12-26) & JP 06 271749 A (POLYPLASTICS CO), 27 September 1994 (1994-09-27) abstract -----	1,27-35

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Information on patent family members

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PCT/US 00/24378

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